

[Amend claim 31 as follows:]

31. (amended) A catalyst corresponding to claim 27, which is characterized by the fact that the metallic oxide(s)  $\text{MO}_3$  which are reduced to  $\text{MO}_2$  are deposited on a support  $\text{TiO}_2$  which itself could be deposited on a substrate having larger surface area than  $\text{TiO}_2$ .

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REMARKS

This application has been amended in a manner that is believed to place it in condition for allowance at the time of the next Official Action.

Claims 27-52 are pending in the present application. Claims 29-31 have been amended to more particularly point out and distinctly claim the present invention. No claims have been canceled. It is believed that no new matter has been added to the present application.

In the outstanding Official Action, claims 29-30 were objected to for reciting informality. The outstanding Official Action suggested that the term "claims" should be amended to to recite "claim". Accordingly, applicants have amended claims 29-30 and thank Examiner Ildebrando for her suggestion for overcoming this objection.

Claims 27-32 were rejected under 35 USC 112, second paragraph, as allegedly being indefinite for failing to

particularly point out and distinctly claim the subject matter which applicant regards as the invention. This rejection is respectfully traversed.

Applicants respectfully submit that the term "MO<sub>2</sub>" type is definite to one of ordinary skill in the art. Moreover, the term is described generally throughout the specification. Specifically, the term is described in the present specification at page 3, lines 32-35 and page 4, lines 5-15.

Claim 31 was rejected for allegedly being indefinite for reciting the recitation "MoO<sub>3</sub>". As suggested by the Examiner, this term has been amended to recite "MO<sub>3</sub>". Once again, applicants thank Examiner Ildebrando for her suggestion as how to overcome this rejection.

In the outstanding Official Action, claims 27-29, 31-37, and 39 were rejected under 35 USC 102(b) as allegedly anticipated by or, in the alternative, under 35 USC 103(a) as obvious over RICHMOND et al. This rejection is respectfully traversed.

It is respectfully submitted that RICHMOND et al. fails to disclose or suggest the present invention. The catalytically active species according to the present invention which are responsible for the hydroisomerization of alkanes are named WO<sub>2</sub> and MoO<sub>2</sub>. Each of these chemical species has bifunctional (metal + acid) properties that are utilized in this catalytic process.

For example, a WO<sub>2</sub> or MoO<sub>2</sub> active phase is obtained, in situ,

by a reduction process using hydrogen for at least 6 hours with  $\text{WO}_3$  at 773 K and with  $\text{MoO}_3$  at 673 K. The dioxides  $\text{WO}_2$  and  $\text{MoO}_2$  are not stable under air or oxygen atmospheres. In such a situation, they undergo oxidation to form catalytically inactive  $\text{WO}_3$  and  $\text{MoO}_3$  states.

Commercial  $\text{WO}_2$  or  $\text{MoO}_2$  samples are therefore usually covered by more than 5 layers of inactive and chemically stable  $\text{WO}_3$  and  $\text{MoO}_3$ . The presence of oxygen or air in the catalytic reactor will lead to the conversion of  $\text{WO}_2$  to  $\text{WO}_3$  and  $\text{MoO}_2$  to  $\text{MoO}_3$ , respectively. If this occurs, the catalytic process will stop.

The goal of using a support in the present invention is to provide a larger surface area of the active phase. A support on which the active  $\text{WO}_2$  or  $\text{MoO}_2$  species could be obtained is  $\text{TiO}_2$ .

The use of other conventional supports such as silica ( $\text{SiO}_2$ ), alumina ( $\text{Al}_2\text{O}_3$ ) or zeolites will lead to the formation of different W or Mo complexes which do not exhibit bifunctional properties and are not active.

The isomerization of alkanes to obtain branched is very difficult (activation energy around 120 kJ/mol) and requires a catalyst having bifunctional properties to perform the dehydrogenation process (metallic function) in order to obtain unsaturated olefins and then to isomerize the olefins on an acidic function (bifunctional mechanism).

Up to now, very expensive Platinum deposited on acidic

alumina or zeolite has been used to complete this isomerization of saturated alkanes.

The isomerization of unsaturated hydrocarbons (olefins) usually takes place on relatively mild acidic sites (most of the oxides with different valances have some acidic properties). These reactions are easier to complete, have relatively low activation energies (less than 60 kJ/mol) and take place at lower reaction temperatures than for the saturated hydrocarbons.

Independent Claim 27 recites a polyvalent bifunctional catalyst, characterized by the fact that it comprises, deposited on a support  $\text{TiO}_2$ , an oxide or a mixture of metallic oxides of  $\text{MO}_2$  type prepared by the reduction of the corresponding  $\text{MO}_3$  oxide (s).

Thus, the claimed invention relates to a bifunctional catalyst capable of isomerizing saturated hydrocarbons (alkanes) compounds. The inventors identified the suboxide,  $\text{MO}_2$  ( $\text{M}=\text{Mo}, \text{W}$ ) which is one of many suboxides  $\text{MO}_x$  which result during the reduction of normally stable  $\text{MO}_3$  to the metal  $\text{M}(0)$  by hydrogen at different temperatures.

The identification of the metallic properties of this  $\text{MO}_2$  phase by many spectroscopic techniques (XPS, UPS...) and the Brönsted acidic properties (by ammonia adsorption) are now well accepted by the scientific community worldwide. Applicants believe that these metallic-acidic properties of this system were not previously known at all.

As explained,  $\text{TiO}_2$  is the support on which it is possible to convert  $\text{MO}_3$ , the initial compound, to  $\text{MO}_2$  by hydrogen reduction at a specific temperature without any negative electronic interaction known as strong metal-support interaction in catalysis which takes place between  $\text{MO}_2$  and the  $\text{TiO}_2$  support surface. In fact, applicants believe that it is not possible to obtain  $\text{MO}_2$  on the surfaces of supports such as alumina, silica or zeolite, precisely because of these electronic interactions. The  $\text{TiO}_2$  support which acts only as a carrier to increase the surface area of the catalyst has no direct catalytic activity itself.

RICHMOND et al. deal with the preparation of diaryl ethers by dehydrating phenols using partially reduced tungsten oxides on different supports.

The catalytic process is dehydration and not dehydrogenation as stated in the applied patent. Moreover, the thermodynamic (energy) requirements are not the same for both processes, demonstrating that the used catalysts cannot be expected to be the same.

The cited oxide in this application is  $\text{WO}_3$  which can be deposited on three different supports: alumina (even preferred), zirconia and titania. It is then reduced by hydrogen at temperatures between  $250 - 450^\circ\text{C}$ . It is believed that as the surface activities (electronic structures) of the three supports are completely different, the nature of the tungsten species present on the surface will be different for each support.

Moreover, it is impossible to totally reduce the tungsten W(VI) to W(IV) under the temperatures of 250-450°C used in this work. This is well known by one skilled in the art.

As a result, applicants believe that RICHMOND et al. could not obtain a bifunctional (metallic-acidic) surface under these experimental conditions. Thus, it is believed that the Official Action fails to disclose or suggest the claimed invention.

The citation of the reduction of W(VI) to W(IV) on page 3, line 40 of RICHMOND et al. is not believed to be relevant to the claimed invention. In fact, this is confirmed by discussing the uncertainty of this assignment by RICHMOND et al.

Thus, applicants believe that it is likely RICHMOND et al. got some oxidic  $W_{20}O_{58}$  (there are many lower valence states than 3 such as  $W_{20}O_{58}$  down to W) with only acidic properties which are typically sufficient for the dehydration of phenols.

Thus, applicants believe that RICHMOND et al. cannot be considered to anticipate or render obvious the present invention.

In the outstanding Official Action, claims 27-29, 31-37 and 39 were rejected under 35 USC 102(b) as allegedly anticipated by or, in the alternative, under 35 USC 103(a) as obvious over YOSHIOKA et al.

Applicants believe that YOSHIOKA et al fail to disclose or suggest the claimed invention. YOSHIOKA et al. relate to the reduction of nitrogen oxides (with ammonia) to nitrogen and

water. The publication describes catalysts comprising oxides or sulfates of selected transition metals of groups: IB, VIB, VB, VIIB and VII of the periodic table and cerium, i.e. oxides from copper, vanadium, chromium, molybdenum, tungsten, manganese, iron and cerium (see column 3, line 64 to column 4, line 3 of said document). However, the oxidation number of the metal oxide is not restricted and oxides of any oxidation number are feasible (see column 4, lines 5-6).

The support used by YOSHIOKA et al. includes a great variety of oxides of titanium such as titanium dioxide ( $\text{TiO}_2$ ), titanium monoxide ( $\text{TiO}$ ), titanium (III) oxide ( $\text{Ti}_2\text{O}_3$ ), ortho and beta titanic acid, and mixtures of these (column 3, lines 36-47).

However, there is no relationship discussed from a catalytic point of view relative to the disclosed oxides of transition metals. Moreover, the species mentioned ( $\text{MoO}_3$ ,  $\text{WO}_3$  and  $\text{WO}_2$ ) are listed without any preference. Applicants clearly believe that this clearly implies that these systems are destined to have the same catalytic properties.

However, as stated above, to get  $\text{WO}_2$ , a reduction process of  $\text{WO}_3$  by hydrogen at 773 K is utilized. Moreover, a characterization method for the identification of this  $\text{WO}_2$  phase has not been identified in YOSHIOKA et al.

While YOSHIOKA et al. have indifferently used several titanium oxides as well as different clays as a support, the chemical and catalytic properties of each of the above mentioned

titanium oxides are completely different. Only  $\text{TiO}_2$  exhibits no catalytic activity. The other oxides negatively interfere with the bifunctional properties of the catalyst itself.

Therefore, it is believed that the work of YOSHIOKA et al. fails to disclose or suggest the claimed invention. As applicants believe that the publication fails to disclose or suggest the bifunctional (metal-acid) properties of  $\text{WO}_2$  produced by hydrogen reduction of  $\text{WO}_3$ , it is believed that YOSHIOKA et al fail to anticipate or render obvious the claimed invention.

In the outstanding Official Action, claims 27-28 and 30-38 are rejected under 35 USC 102(b) as allegedly anticipated by or, in the alternative, under 35 USC 103(a) as obvious over MIKI et al.

The MIKI et al. publication is directed to the production of phenol from benzoic acid using as a catalyst a precise combination comprising not less than at least 4 active species, namely an iron oxide, a nickel oxide, a vanadium/molybdenum oxide and an alkali metal/alkaline earth metal oxide (see abstract or claim 1 of this application).

Preferably, the molybdenum oxide is molybdenum trioxide  $\text{MoO}_3$ , although it may include molybdenum dioxide  $\text{MoO}_2$  (see page 3, line 40 of said document). However, it appears that none of the numerous examples given (almost 200!) mention the presence of the active dioxide. The catalyst of this publication may indifferently be supported on titanium oxide or silica (see page



4, lines 13-14). Oxygen is supplied along with the raw material (benzoic acid) for the conversion process excluding therewith any hypothetical possibility of reduction of the catalytic oxides (see page 4, lines 47-56).

Since there are no similarities between the chemical and catalytic properties of the different nickel, iron, vanadium and molybdenum oxides and those of  $\text{MoO}_2$  or  $\text{WO}_2$ , these oxides cannot perform the isomerization of alkanes. MIKI et al. did not reveal the claimed bifunctionality of  $\text{MoO}_2$  nor its preparation and deposition on the only available support ( $\text{TiO}_2$ ) on which this bifunctionality is preserved.

Even if one skilled in the art contemplated a preparation of  $\text{MoO}_2$  which starts from  $\text{MoO}_3$  (as it is supposed to be), such a preparation would not be possible with the teachings of this document because of the presence of oxygen in the reaction mixture (page 4, lines 47-55). Therefore, applicant believes that the publication fails to satisfy the recitation of the claimed invention.

Moreover, applicants believe that the possible use of silica as a support in this document is a clear indication that the disclosed catalytic system cannot be related to a catalyst according to the present invention.

Thus, applicants respectfully submit it is believed that MIKI et al. fail to disclose or suggest the claimed invention.

At this time, applicants respectfully request the rejoinder of claims 40-52. In the outstanding Official Action, the Examiner stated that "applicant must demonstrate that the special technical feature defines the contribution of the prior art in order to argue that there is unity of invention". Applicants respectfully traverse this assertion.

The same claims as pending in the present national stage application were subject to examination during the international phase in the PCT application.

It is believed that that the Official Action fails to comply with the requirements of PCT Rules 13.1 and 13.2, in seeking to justify the lack of unity determination. Specifically, the definition of "special technical feature" in PCT Rule 13.2 is art-based. Therefore, a proper lack of unity determination requires citation of a reference showing the lack of unity of an invention. This burden is on the Patent Office and not the applicant.

As noted above, RICHMOND et al. (EPO 0584415) fail to disclose or suggest the claimed invention. As a result, it is believed the Official Action fails to comply with the requirements of PCT Rules 13.1 and 13.2. Because the Official Action fails to comply with the requirements of PCT Rules 13.1 and 13.2, it is believed that the Official Action fails to provide a proper lack of unity determination. Absent a proper citation having been made, the lack of unity determination is

believed to be improper as a matter of law.

With this in mind, applicants note that the international Examiner found no lack of unity, applying the same legal standard to the identical facts. Moreover, it is believed the Official Action does not explain why, applying the identical legal standards to the identical claims, the opposite result is now being reached in the present U.S. national phase application, relative to the international application.

Thus, in view of the above, applicants respectfully request the rejoinder of claims 40-52.

In view of the present amendment and the foregoing remarks, therefore, it is believed that the present application has been placed in condition for allowance, with claims 27-52, as presented. Allowance and passage to issue on that basis are accordingly respectfully requested.

Attached hereto is a marked-up version of the changes made to the claims. The attached page is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE."

Respectfully submitted,

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By



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**VERSION WITH MARKINGS TO SHOW CHANGES MADE**

IN THE CLAIMS:

Claim 29 has been amended as follows:

29. (amended) A catalyst corresponding to [claims] claim 27, characterized by the fact that the metallic oxide obtained by reduction on the support is  $\text{WO}_2$ .

Claim 30 has been amended as follows:

30. (amended) A catalyst corresponding to [claims] claim 27, characterized by the fact that the metallic oxide obtained by reduction on the support is  $\text{MoO}_2$ .

Claim 31 has been amended as follows:

31. (amended) A catalyst corresponding to claim 27, which is characterized by the fact that the metallic oxide(s) [ $\text{MoO}_3$ ]  $\text{MO}_3$  which are reduced to  $\text{MO}_2$  are deposited on a support  $\text{TiO}_2$  which itself could be deposited on a substrate having larger surface area than  $\text{TiO}_2$ .